Effect of Al mole fraction on carrier diffusion lengths and lifetimes in $Al_xGa_{1-x}As$

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The ambipolar diffusion length and carrier lifetime are measured in $Al_xGa_{1-x}As$ for several mole fractions in the interval $0 < x < 0.38$. These parameters are found to have significantly higher values in the higher mole fraction samples. These increases are attributed to occupation of states in the indirect valleys, and supporting calculations are presented.

Measurement of certain basic material parameters such as carrier diffusion length, lifetime, and mobility is important for device modeling and material characterization. The $Al_xGa_{1-x}As$ system is an important material system for photonic applications such as lasers, light-emitting diodes (LEDs), and photovoltaic devices, as well as electronic devices such as high electron mobility transistors and resonant tunneling diodes. Although the electron mobility as a function of Al mole fraction has been studied systematically,[1] reports of lifetimes or diffusion lengths in $Al_xGa_{1-x}As$ are scattered[2,3] and performed at lower excitation levels than those used in LEDs and lasers. In this letter, we report on measurements of the ambipolar diffusion length and carrier lifetime in $Al_xGa_{1-x}As$ for several values of x in the interval $0 < x \leqslant 0.38$. Both the diffusion length and the lifetime show a dramatic increase for Al mole fraction near 0.4. Based on a consideration of the relative electron densities in the direct and indirect valleys, the increase in lifetime is attributed to population of the indirect valleys for $x \approx 0.4$.

The samples used in this experiment were grown by molecular beam epitaxy in a Riber 2300 R&D system. Layers with Al mole fractions of 0.09, 0.19, 0.25, 0.35, and 0.38 were grown as well as a GaAs layer and a 100 Å GaAs quantum well. There was no intentional doping in any of the samples. The determination of the Al mole fraction was accomplished by room-temperature photoluminescence (PL) measurements. The PL spectra were taken using a relatively low intensity (≈ 1 W/cm^2) excitation by the 5145 Å line of an argon ion laser, and Al mole fractions x were determined from the PL spectra peak energy E_p, using the relation $E_p\,(\text{eV}) = 1.42 + 1.45x - 0.25x^2$, which has been shown to hold for $0 < x \leqslant 0.45$ and similar excitation conditions.[4] There is some disagreement as to the relationship between band-gap energy and Al mole fraction,[5,6] particularly in the range $0.35 < x < 0.45$. We have used the relationship of Ref. 6 since it is more recent.

Determination of the ambipolar diffusion length was accomplished by a cathodoluminescence (CL) technique described elsewhere.[7] The experiment is performed in a modified scanning electron microscope (SEM) with a fiber optic cathodoluminescence (CL) collection system.[8] In the SEM, an energetic electron beam is incident upon the sample, generating electron-hole pairs within the interaction region of the beam with the sample. A thin aluminum mask covers half of the sample and prevents detection of the luminescence emanating from the region under it. All detected luminescence is from radiative recombination which occurs in the unmasked region. By generating the carriers in the masked region of the sample and measuring the luminescence intensity as a function of the distance of the beam from the mask edge, we are able to determine the diffusion length. To allow different material compositions to be investigated on a single sample, carriers were confined to narrow channels of the material being studied by high band-gap barriers on either side. Signals from different channels were at different wavelengths and were separated with a monochromator.

Figure 1 shows the results of the diffusion length measurements. These data were taken at room temperature at carrier densities in the range 10^{17}–10^{18} cm^{-3}. For an Al mole fraction <0.3 the diffusion length remains fairly constant, but for larger values of x it shows a marked increase. The increase in the diffusion length implies an increase in either mobility or lifetime since $L_D = \sqrt{D\tau} = \sqrt{k_B T \mu \tau / q}$, where L_D, D, and μ are the ambipolar diffusion length, diffusivity, and mobility and where τ, k_B, T, and q are the carrier lifetime, Boltzmann's constant, temperature, and the electron charge, respectively. It is not unreasonable to expect the lifetime to increase as x approaches 0.4 since this is near the point at which AlGaAs becomes an indirect gap semiconductor and lifetimes in the indirect valleys can be quite long if the material is of good quality. Using the electron mobility data of Ref. 1, and a heavy hole mobility calculated from

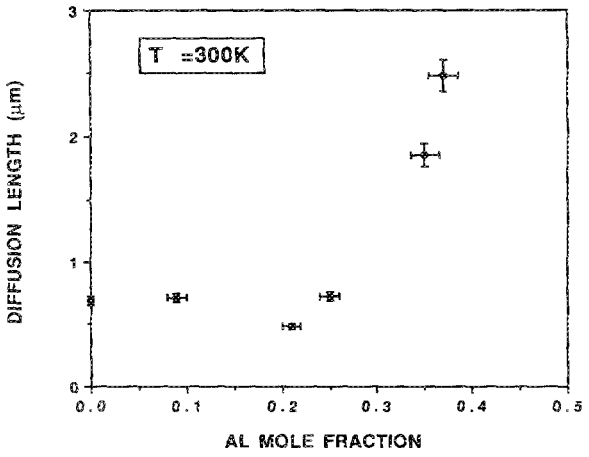

FIG. 1. Ambipolar diffusion length at a carrier density of 10^{17}–10^{18} cm^{-3}. The samples with Al mole fraction >0.3 have high values due to the increased lifetime. A 100 Å GaAs quantum well showed the same value as the thicker GaAs ($x = 0.0$) layer.

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Ref. 9, our L_D measurements imply an order of magnitude increase in lifetime for the 0.35 and 0.38 Al mole fraction samples as compared to GaAs.

To confirm this interpretation, carrier lifetimes were measured by examining the photoluminescence decay signal produced by short-pulse optical excitation of the sample. In this experiment, the sample was excited using frequency-doubled pulses from a mode-locked Nd:YAG laser. The laser pulses typically had a 100 ps width and a peak power of 1 W. An electro-optic pulse extractor reduced the repetition rate from 100 MHz to 10 kHz. The luminescence was coupled into a monochromator which acted as a bandpass filter, passing only the luminescence from the region of interest. The luminescence decay was measured with a photomultiplier tube and a boxcar integrator. The system response was measured to be 4 ns.

The results of the lifetime measurement are summarized in Fig. 2. These data were taken at a carrier density of $\sim 3\times 10^{18}$ cm^{-3}. The samples with Al mole fraction <0.3 all showed a lifetime at the system limit of 4 ns. Reports on GaAs lifetimes for this carrier density are in the 2–4 ns range.[5,10] Lifetimes on the 0.35 and 0.38 mole fraction samples are an order of magnitude higher than this, in agreement with the predictions from the diffusion length measurements. These samples also showed a trend in increasing lifetime as the pumping power was increased in apparent contradiction with expectations for a bimolecular recombination process. These effects can be explained by examining the relative populations of the direct and indirect bands.

As the Al mole fraction increases from zero to 0.35, the energy separation between the Γ, X, and L valleys decreases to <0.05 eV.[5,6] This situation persists over the range $0.35<x<0.45$. In this region, all three valleys will be significantly populated under room-temperature excitation. Since intervalley scattering and intraband relaxation processes all occur on a time scale which is much shorter than the carrier lifetime,[11,12] the carriers in the conduction band may be assumed to be in thermal equilibrium, and Fermi–Dirac statistics may be applied. We have calculated the populations of the three valleys of the conduction band using Fermi–Dirac statistics and the parabolic band approximation. The band gaps for these valleys were taken from Ref. 6 and the density-of-states effective masses from Ref. 9. We define the parameter $R(n)\equiv n/n_\Gamma$, where n is the carrier concentration and n_Γ is the population of the Γ valley. On average, each electron will spend $1/R$ of its life in the Γ valley. Taking the lifetime in the indirect valleys as infinite compared to the lifetime in the Γ valley, R can be viewed as a lifetime enhancement factor; that is, the carrier lifetime is given by $\tau = R\tau_\Gamma$, where τ_Γ is the lifetime in the Γ valley. In Fig. 3, $R(n)$ is plotted for Al mole fractions of 0.25, 0.35, and 0.38. At low carrier concentrations, the curves are flat since, in this region, the Fermi level is in the gap and the Fermi–Dirac distribution above the band edge is Boltzmann-like, leading to relative populations that are approximately independent of carrier density. For example, the relative population of the Γ and X valleys in this region is given approximately by $n_X/n_\Gamma = (m_X/m_\Gamma)^{3/2}e^{-\Delta E/k_BT}$, where m_X and m_Γ are the density-of-states effective masses in the X and Γ valleys, respectively, and ΔE is the energy separation between the X and Γ band minima. As the Fermi level rises to within $2k_BT$ of the indirect valleys, R begins to increase sharply. This explains our observations of longer lifetimes for higher pump powers. Eventually, R begins to level off and approaches the value $(m_X+m_L+m_\Gamma)/m_\Gamma$ asymptotically. Although it is questionable whether these high carrier concentrations could be achieved without other effects dominating, values of $R\geqslant 35$ should be attainable at lower carrier concentrations for slightly higher Al mole fractions. To compare these calculations with our data, we assume a value for τ_Γ of 3 ns. At a carrier concentration comparable to our experiments, this predicts lifetimes of 32, 12, and 3 ns, in good agreement with our measured values.

In conclusion, we have presented measurements of ambipolar diffusion lengths and carrier lifetimes in $Al_xGa_{1-x}As$ for several Al mole fractions in the interval

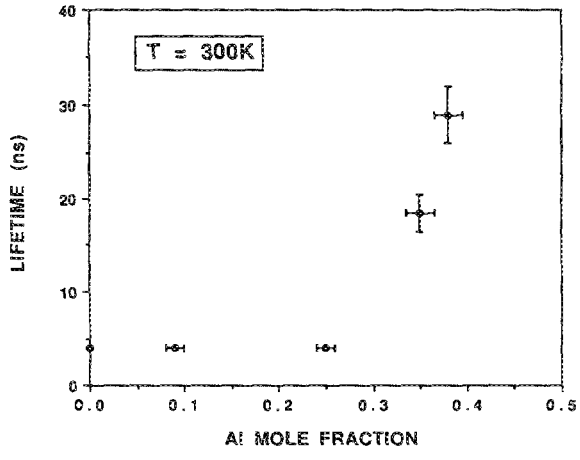

FIG. 2. Carrier lifetimes at a carrier density of $\sim 3\times 10^{18}$ cm^{-3}. The long lifetimes for the 0.35 and 0.38 mole fraction samples are attributed to the population of the X and L valleys. The values for the samples with Al mole fraction <0.3 are upper limits since the system response was 4 ns.

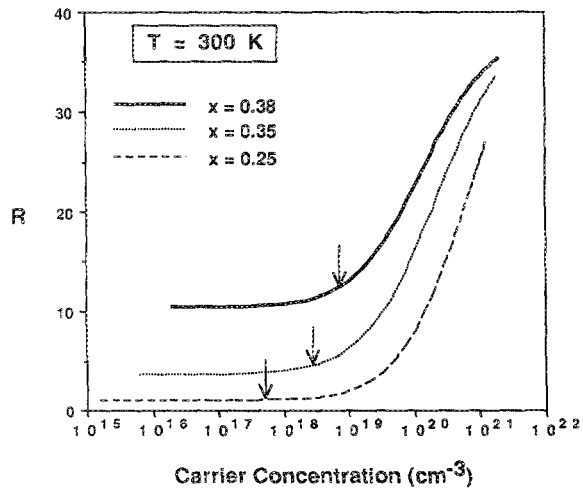

FIG. 3. Ratio of the carrier concentration to the population of the Γ valley as a function of carrier concentration for three different Al mole fractions. The arrows show the concentration at which the quasi-Fermi level is at the bottom of the conduction band. The lower and upper endpoints of the curves correspond to quasi-Fermi levels 0.15 eV below and 0.55 eV above the conduction-band edge.

$0 < x < 0.38$. Samples having compositions near the direct gap–indirect gap transition show significantly increased lifetimes and diffusion lengths. We presented calculations indicating that, at these mole fractions, carriers are occupying states in the X and L valleys for a substantial fraction of their lifetime. These calculations indicate that the lifetime may be as long as 100 ns with slightly higher mole fractions than investigated here. Measurements on the temperature dependence of the lifetime may prove useful in determining the composition at which $Al_xGa_{1-x}As$ becomes an indirect gap semiconductor.

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